

# Gage-equivalent forms of the Schrödinger equation for a hydrogenlike atom in a time-dependent electric field

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## Abstract

Several gage-equivalent forms (including some novel ones) of the Schrödinger equation for a hydrogenlike atom in a time-dependent electric field of a laser pulse are presented. These forms allow to develop a perturbation theory for both small and rather large intensities of the electromagnetic field.

## 1 Introduction

Exploring the interaction between the electromagnetic field and matter is the major and oldest issue of both classical and quantum physics. It has a number of branches. One of them is the interaction of a strong laser field with atoms and molecules. Here the concept of a “strong field” implies the field which is comparable with an electric field in an atom, which binds electrons to nucleus. The strong field may result in single or multiple ionization of an atom or molecule. By studying the energy and angular distributions of the

ionized electrons physicists expect to obtain information about the structure of a quantum object and the ionization mechanisms.

The coming into being of the theory of such processes is due to pioneering works of Keldysh [1] and his followers. Since then, a number of studies have appeared analyzing pluses and minuses of this theory, investigating its limits and suggesting corrections for its improvement. Just citing these works would take a half of the journal volume and therefore we cite only several recent review papers [2, 3, 4, 5, 6, 7], where trends in this field of science can be seen. However, the theoretical and mathematical content of the majority of works in recent years has shifted towards development of numerical schemes for solving basic equations. This is due to the intense growth of computational facilities of modern computers. Despite the enormous progress of such approach in understanding the processes taking place under the action of an intense laser pulse, the analytical models remain of very current importance, since they have the power of prediction. The exact solution of the considered problem is known only for a very small set of local potentials, in particular, for an oscillating potential (see, for instance, [8, 9]). However, there is no ionized states in this potential. For the simplest practical case, namely for the hydrogen atom, one already must consider different approximations whose mathematical correctness is not always clear.

In this connection the property of gauge invariance of the electromagnetic field is often helpful. In turn, this property allows to obtain various equivalent forms of the time-dependent Schrödinger equation (TDSE) related to each other through unitary transformations which, as is known, lead to the invariance of the physical quantities given by quadratic forms of the wave function. Let us recall that the Maxwell equations can be written in terms of the scalar and vector potentials,  $U(\vec{r}, t)$  and  $\vec{A}(\vec{r}, t)$ . These potentials quite unambiguously determine the observed characteristics of the electromagnetic field, namely the electric and magnetic field intensities,  $\vec{E}$  and  $\vec{H}$ . At that, the potentials themselves are defined ambiguously. For example, two sets of potentials  $(\vec{A}', U')$  and  $(\vec{A}, U)$ , where

$$\vec{A}' = \vec{A} + \vec{\nabla}f, \quad U' = U - \frac{1}{c} \frac{\partial f}{\partial t},$$

give the same electric and magnetic field intensities for an arbitrary function  $f(\vec{r}, t)$ .

The forms of TDSE using different gauge transformations of the electromagnetic field and some of the corresponding useful consequences are the

subject of this work. The atomic units  $e = m_e = \hbar = 1$  are used throughout. According to this system of units the light velocity  $c$  is about 137.

## 2 Coordinate representation

First of all it should be noted that the field intensity is, upon definition, related to the potential as follows:

$$\vec{E}(\vec{r}, t) = -\frac{1}{c} \frac{\partial}{\partial t} \vec{A}(\vec{r}, t) - \vec{\nabla} U(\vec{r}, t). \quad (1)$$

In the so-called Coulomb gage it is assumed that

$$\text{div} \vec{A} = 0.$$

In the simplest study one makes a physical assumption about a weak dependence of the scalar potential on the coordinate within the atom, i.e.

$$U(\vec{r}, t) \simeq U(0, t),$$

which allows to neglect the gradient of the scalar potential in (1). This leads to a well known dipole approximation

$$\vec{A}(\vec{r}, t) \simeq \vec{A}(0, t) = \vec{A}(t).$$

Setting  $\vec{A}(t) = \vec{e} A(t)$ , where  $\vec{e}$  is the unit polarization vector, we obtain a linearly polarized laser beam. The condition of the absence of the field outside the time interval  $(0, T)$ , where the laser pulse acts, takes the form  $A(t \leq 0) = A(t \geq T) = 0$ .

Consider the TDSE, which describes the interaction between an electric pulse and a hydrogenlike atom,

$$\left\{ i \frac{\partial}{\partial t} - \frac{1}{2} \left[ -i \vec{\nabla} + \frac{1}{c} \vec{e} A(t) \right]^2 + \frac{Z}{r} \right\} \Psi(\vec{r}, t) = 0, \quad \Psi(\vec{r}, 0) = \sqrt{\frac{Z^3}{\pi}} e^{-Zr}, \quad (2)$$

where  $Z$  designates the nuclear charge. The initial state of the problem allows to conclude that at any time moment  $t$  we deal with a square integrable wave packet. Moreover, a normalization condition should be fulfilled:

$$\int d\vec{r} |\Psi(\vec{r}, t)|^2 = 1, \quad (3)$$

whose physical meaning is a conservation of the total probability of all events in the system.

The well known unitary transformation

$$\Psi(\vec{r}, t) = \exp \left[ -i \frac{1}{c} A(t) (\vec{e}\vec{r}) \right] \Phi_L(\vec{r}, t)$$

results in the following form of TDSE:

$$\left[ i \frac{\partial}{\partial t} + \frac{1}{2} \Delta - \vec{E}(t) \vec{r} + \frac{Z}{r} \right] \Phi_L(\vec{r}, t) = 0, \quad \Phi_L(\vec{r}, 0) = \Psi(\vec{r}, 0). \quad (4)$$

Here  $\vec{E}(t) = -\vec{e} \partial A(t) / c \partial t$ . The notation  $\Phi_L(\vec{r}, t)$  indicates the so-called length form of TDSE. In this context,  $\Psi(\vec{r}, t) \equiv \Phi_V(\vec{r}, t)$  is sometimes referred to as the velocity form. Usually one requires a good numerical algorithm to give a coincidence (within the accuracy) of the computed observed quantities (the level occupations, angular and energy distributions of the ionized electrons and etc.) in the length and velocity forms. In the exact theory they must be identical.

The less known Henneberger-Kramers transformation employs the unitary operator of the space shift [10]:

$$\Psi(\vec{r}, t) = \exp \left[ b(t) (\vec{e}\vec{\nabla}) - \frac{i}{2c^2} \int_0^t A^2(\tau) d\tau \right] \Phi_{HK}(\vec{r}, t), \quad b(t) = -\frac{1}{c} \int_0^t A(\tau) d\tau. \quad (5)$$

Inserting (5) into (2) and setting for convenience that  $b(t) = Af(t)$ , where  $|f(t)| \leq 1$  and  $f(t \leq 0) = f(t \geq T) = 0$ , we get

$$\left[ i \frac{\partial}{\partial t} + \frac{1}{2} \Delta + \frac{Z}{|\vec{r} - \vec{e} A f(t)|} \right] \Phi_{HK}(\vec{r}, t) = 0, \quad \Phi_{HK}(\vec{r}, 0) = \Psi(\vec{r}, 0). \quad (6)$$

Note that at any time moment  $t$  the wave packet is normalized to unity:

$$\int d\vec{r} |\Phi_{HK}(\vec{r}, t)|^2 = 1.$$

Making the following scaling transformation:

$$t = A\tau, \quad \vec{r} = A\vec{x}, \quad \Phi_{HK}(\vec{r}, t) = A^{-3/2} \phi(\vec{x}, \tau),$$

we obtain

$$\left[ iA \frac{\partial}{\partial \tau} + \frac{1}{2} \Delta_x + \frac{AZ}{|\vec{x} - \vec{e}f(A\tau)|} \right] \phi(\vec{x}, \tau) = 0. \quad (7)$$

Let us set

$$\phi(\vec{x}, \tau) = N e^{-AS(\vec{x}, \tau)}, \quad (8)$$

where  $\Re(S) > 0$  if  $x \rightarrow \infty$ . It follows from (8) that

$$\left[ i \frac{\partial S}{\partial \tau} - \frac{1}{2} (\vec{\nabla}_x S)^2 \right] + \frac{1}{A} \left[ \frac{1}{2} \Delta_x S - \frac{Z}{|\vec{x} - \vec{e}f(A\tau)|} \right] = 0. \quad (9)$$

In the absence of an external electric field,  $f(t) = 0$  and

$$S_0(\vec{x}, \tau) = Zx - \frac{i}{2} Z^2 \tau.$$

This function satisfies not only (9) but also the equation

$$\left[ i \frac{\partial S_0}{\partial \tau} - \frac{1}{2} (\vec{\nabla}_x S_0)^2 \right] = 0.$$

This fact allows to use the perturbation series if  $A \gg 1$ :

$$S(\vec{x}, \tau) = \sum_{n=0} \left( \frac{1}{A} \right)^n S_n(\vec{x}, \tau). \quad (10)$$

For instance, the term  $S_1$  satisfies the linear nonhomogeneous partial differential equation

$$i \frac{\partial S_1}{\partial \tau} - \left( \frac{\vec{x}}{x} \vec{\nabla}_x \right) S_1 + \left[ \frac{Z}{x} - \frac{Z}{|\vec{x} - \vec{e}f(A\tau)|} \right] = 0, \quad (11)$$

whose particular solution is

$$S_1(\vec{x}, \tau) = i \int_0^\tau d\xi \left[ \frac{Z}{i\xi + C} - \frac{Z}{|(i\xi + C)\vec{x}/x - \vec{e}f(A\xi)|} \right]. \quad (12)$$

In formula (12),  $C = x - i\tau$  is an integral of motion of equation (11), in which connection

$$|(i\xi + C)\vec{x}/x - \vec{e}f(A\xi)| = \sqrt{(i\xi + C)^2 + f^2(A\xi) - 2 \frac{(\vec{x}\vec{e})}{x} (i\xi + C) f(A\xi)}.$$

After the termination of the laser pulse ( $t \geq T$ )

$$S_1(\vec{x}, \tau) = i \int_0^{T/A} d\xi \left[ \frac{Z}{i\xi + C} - \frac{Z}{|(i\xi + C)\vec{x}/x - \vec{e}f(A\xi)|} \right]. \quad (13)$$

For the term  $S_2$  we have, in accordance with (9) and (10), the equation

$$\left[ i \frac{\partial S_2}{\partial \tau} - \left( \frac{\vec{x}}{x} \vec{\nabla}_x \right) S_2 \right] + \frac{1}{2} [\Delta_x S_1 - (\vec{\nabla}_x S_1)^2] = 0. \quad (14)$$

Correspondingly, its solution is

$$S_2(\vec{x}, \tau) = \frac{i}{2} \int_0^\tau d\eta [\Delta_x S_1 - (\vec{\nabla}_x S_1)^2]. \quad (15)$$

To apply the gradient and Laplas operators to the function  $S_1(\vec{x}, \eta)$  in (15), at first it is necessary to set  $C = x - i\tau$  in (12), then to perform these differential operations, and after that to make the substitution  $\vec{x} = (i\eta + C)\vec{x}/x$ .

In (13) and (15) one can return back to the variables  $(\vec{r}, t)$  and see that the argument of the exponent in (8) does not explicitly depend on  $A$ . This is a footprint of a quasi-classical approximation.

For estimating the value of  $A$  we consider a particular case of the laser pulse shape which is frequently utilized in calculations:

$$A(t) = \begin{cases} A_0 \sin^2(\pi t/T) \sin(\omega t + \varphi) & (0 \leq t \leq T), \\ A(t) = 0 & (t \geq T), \end{cases} \quad \frac{A_0}{c} = \frac{1}{\omega} \sqrt{\frac{I}{I_0}}. \quad (16)$$

In (16),  $I_0 = 3.5 \times 10^{16}$  Wt/cm<sup>2</sup> is the unit of the field intensity in an atom,  $\omega = 0.056$  (the base frequency of the titan-sapphire laser),  $T \approx 2\pi n/\omega$ , and  $n$  is a number of cycles in the pulse. The phase  $\varphi$  must be chosen according to the condition  $b(T) = 0$ . Setting  $n = 10$  and  $I \sim 10^{14}$  Wt/cm<sup>2</sup>, we obtain the estimate  $A \sim 20$ , or  $1/A \sim 0.05$ . This allows to expect a good convergence of the series (10) in the case of a rather strong field with moderate carrier frequency. In this range the experimental data have been obtained which allow to check the correctness of the derived expansion in the reversed powers of the field intensity.

### 3 Momentum representation

TDSE for the considered problem in momentum space follows from (2) and has the form

$$\left\{ i \frac{\partial}{\partial t} - \frac{1}{2} \left[ \vec{p} + \frac{1}{c} A(t) \vec{e} \right]^2 \right\} \tilde{\Psi}(\vec{p}, t) + \int \frac{d^3 p'}{(2\pi)^3} \frac{4\pi Z}{|\vec{p} - \vec{p}'|^2} \tilde{\Psi}(\vec{p}', t) = 0, \quad (17)$$

$$\tilde{\Psi}(\vec{p}, 0) = \frac{8\sqrt{\pi Z^5}}{(p^2 + Z^2)^2}.$$

In equation (17) the function  $\tilde{\Psi}(\vec{p}, t)$  designates the Fourier transform of the function  $\Psi(\vec{r}, t)$ . The unitary transformation

$$\tilde{\varphi}(\vec{p}, t) = e^{i \frac{t}{2} p^2 - i b(t) (\vec{e} \vec{p}) + i \frac{1}{2} \int_0^t d\tau [b'(\tau)]^2} \tilde{\Psi}(\vec{p}, t) \quad (18)$$

leads to the equation

$$i \frac{\partial}{\partial t} \tilde{\varphi}(\vec{p}, t) + \frac{Z}{2\pi^2} \int \frac{d^3 x}{x^2} e^{-i \frac{t}{2} x^2 + i [t\vec{p} - b(t)\vec{e}] \vec{x}} \tilde{\varphi}(\vec{p} - \vec{x}, t) = 0. \quad (19)$$

One can obtain the analogous equation in coordinate space upon making the Fourier transform

$$\tilde{\varphi}(\vec{p}, t) = \int d^3 r \, e^{-i \vec{p} \vec{r}} \varphi(\vec{r}, t).$$

In this case we obtain from (19)

$$i \frac{\partial}{\partial t} \varphi(\vec{r}, t) + \frac{Z}{2\pi^2} \int \frac{d^3 x}{x^2} e^{i \frac{t}{2} x^2 + i [\vec{r} - b(t)\vec{e}] \vec{x}} \varphi(\vec{r} + t\vec{x}, t) = 0. \quad (20)$$

Using the momentum shift operator, equation (19) can be presented in the form

$$i \frac{\partial}{\partial t} \tilde{\varphi}(\vec{p}, t) + \frac{Z}{2\pi^2} \int \frac{d^3 x}{x^2} e^{-i \frac{t}{2} x^2 + i [t\vec{p} - b(t)\vec{e}] \vec{x}} e^{-\vec{x} \vec{\nabla}_p} \tilde{\varphi}(\vec{p}, t) = 0. \quad (21)$$

The Weyl operator identity leads to the following result:

$$e^{i \vec{p} \vec{x} t} e^{-\vec{x} \vec{\nabla}_p} \equiv e^{i \frac{t}{2} x^2} e^{i \vec{x} (\vec{p} t + i \vec{\nabla}_p)},$$

which allows to obtain from (21) the equation

$$i \frac{\partial}{\partial t} \tilde{\varphi}(\vec{p}, t) + \frac{Z}{2\pi^2} \int \frac{d^3 x}{x^2} e^{i \vec{x} \vec{H}} \tilde{\varphi}(\vec{p}, t) = 0. \quad (22)$$

Here  $\vec{\mathcal{H}} = t\vec{p} - b(t)\vec{e} + i\vec{\nabla}_p$ . Equation (22) can be presented in a more compact operator form, upon integrating over  $\vec{x}$ :

$$i\frac{\partial}{\partial t}\tilde{\varphi}(\vec{p}, t) + \frac{Z}{|t\vec{p} - b(t)\vec{e} + i\vec{\nabla}_p|}\tilde{\varphi}(\vec{p}, t) = 0, \quad \tilde{\varphi}(\vec{p}, 0) = \tilde{\Psi}(\vec{p}, 0). \quad (23)$$

A similar equation can be obtained for  $\varphi(\vec{r}, t)$  as well:

$$i\frac{\partial}{\partial t}\varphi(\vec{r}, t) + \frac{Z}{|\vec{r} - b(t)\vec{e} - it\vec{\nabla}_r|}\varphi(\vec{r}, t) = 0, \quad \varphi(\vec{r}, 0) = \Psi(\vec{r}, 0). \quad (24)$$

The eigenfunctions of the operator  $\vec{\mathcal{H}}$  are the Volkov states [11]

$$\chi(\vec{p}, t) = e^{i\frac{t}{2}p^2 - ib(t)(\vec{e}\vec{p}) - i\vec{p}\vec{r}},$$

i.e.  $f(\vec{\mathcal{H}})\chi = f(\vec{r})\chi$ . Expanding the function  $\tilde{\varphi}(\vec{p}, t)$  over the basis of the Volkov states, we again obtain (17).

Thus, we arrive at the operator equation

$$\frac{\partial \mathcal{S}(t)}{\partial t} = iZ\mathcal{A}(t)\mathcal{S}(t), \quad \mathcal{S}(0) = I, \quad \tilde{\varphi}(\vec{p}, t) = \mathcal{S}(t)\tilde{\varphi}(\vec{p}, 0), \quad (25)$$

with  $\mathcal{A}(\tau) = 1/|\vec{\mathcal{H}}|$ . Its formal solution can be presented, for example, in the form of the Magnus expansion [12]

$$\mathcal{S}(t) = \exp \left[ iZ \int_0^t d\tau \mathcal{A}(\tau) \right] = \exp \left( \sum_{n=1}^{\infty} B_n \right), \quad (26)$$

where

$$\begin{aligned} B_1 &= iZ \int_0^t d\tau \mathcal{A}(\tau), \\ B_2 &= -\frac{Z^2}{2!} \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 [\mathcal{A}(\tau_1), \mathcal{A}(\tau_2)], \\ B_3 &= -\frac{iZ^3}{3!} \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \int_0^{\tau_2} d\tau_3 \{ [\mathcal{A}(\tau_1), [\mathcal{A}(\tau_2), \mathcal{A}(\tau_3)]] + [[\mathcal{A}(\tau_1), \mathcal{A}(\tau_2)], \mathcal{A}(\tau_3)] \} \end{aligned}$$

and so on. This leads to necessity of calculating commutators of the operator  $\mathcal{A}(t)$  at different time moments  $t$ .



Despite the aesthetic attraction and symmetry of equations in the  $(\vec{r}, \vec{p})$  variables, one can use it only in the context of the perturbation theory with respect to the reverse powers of the parameter  $A$  (see (6)). One can obtain the exact solutions of equation (25), if the operator  $\mathcal{A}(t)$  is expressed as a finite linear combination of the Lie algebra generators with time-dependent coefficients. As to the solution in the form of the Magnus expansion (26), its utilization seems to be not very efficient in practice due to unclear physical interpretation of the operators  $\exp(B_i)$ , as opposed, for example, to the cases of the space and momentum shift operators.

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